# Towards the next generation of highefficiency Cu(In,Ga)Se<sub>2</sub> thin-film solar cells – Sharc25

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#### Abstract

The EU Horizon Sharc25 project has provided deep insights into highly efficient Cu(In,Ga)Se, (CIGSe) thin-film solar cells fabricated by lowand high-temperature co-evaporation using advanced characterization methods, analytical tools, device simulation, and density functional theory modelling. This complementary approach led to a continuous knowledgedriven development and improvement of the CIGSe absorber. Based on optimized chemical composition, profiles, and alkali metal post-deposition treatments (PDT) using KF, RbF, and CsF, the CIGSe cell efficiency could be substantially increased to a record value of 22.6%. Due to additional modifications at the absorber/emitter (replacement of standard buffer system by a combination of thin CdS and TiO<sub>2</sub>) and back contact/ absorber (introduction of Al back reflector in combination with InZnO diffusion barrier) interfaces, in particular the short-circuit current could be increased. Furthermore, passivation layers in combination with point contact schemes at the CIGSe front and back side were developed and are still under investigation.

#### Introduction

Over the last five years, many research institutes and companies have increased their efficiency values for chalcopyrite-type thin-film solar cells to a level of close to 20% or even above, as listed in Table 1. These high values could be reached with Cu(In,Ga)Se<sub>2</sub> (CIGSe), (Ag,Cu)(In,Ga)Se<sub>2</sub> (AgCIGSe), and Cu(In,Ga)(S,Se)<sub>2</sub> (CIGSSe) absorber layers grown by the co-evaporation (simultaneous evaporation of all elements) or sequential deposition process (deposition of metallic precursors and subsequent selenization and/or sulfurization).

#### The Sharc25 project: an overview

Comparing the high efficiencies for chalcopyritebased solar cells achieved by several groups worldwide, as described in Table 1, with the theoretical Shockley-Queisser limit of 33% for single-junction solar cells, there is still a large gap of >10% (absolute) between the experimental results and the maximum theoretical efficiency. The efficiency gap is a result of differences between all experimental solar cell parameters such as open-circuit voltage  $(V_{\rm oc})$ , short-circuit current density  $(J_{sc})$ , and fill factor (*FF*) compared to their maximum theoretical counterparts. This difference was the main reason why the project "Sharc25" was initiated in 2014 and started in 2015 within the EU Horizon 2020 programme. The goal of Sharc25 is to challenge the key limiting factors in state-of-the-art CIGSe solar cells, namely non-radiative carrier recombination and light absorption losses in emitter layers. Electronic losses like recombination at the absorber/buffer interface and at the back contact are addressed. Different kinds of fluctuations and defects within the absorber are identified and addressed as well as optical losses such as reflection at and absorption in the transparent conductive oxide (TCO) emitter, reflection and absorption in the buffer layer, and insufficient absorption/recombination in the CIGSe absorber. The acronym "Sharc25" stands for "Super high efficiency Cu(In,Ga)Se, thin-film solar cells approaching 25%".

The Sharc25 project is coordinated by the Centre for Solar Energy and Hydrogen Research Baden-Württemberg (ZSW) and the consortium consists of 11 partners from eight European countries including R&D partners, universities, national laboratories, and two companies (Flisom AG and NICE Solar Energy GmbH). The task of the companies is to assess the experimental and theoretical results of the consortium with respect to their relevance to and compatibility with industrial large-scale mass production. The idea behind the project is to pool the complementary multidisciplinary skills of the involved research groups in a bid to push the CIGSe single junction solar cell performance towards the theoretical limit, as illustrated in Figure 1. The Sharc25 project started in May 2015 and will run until the end of October 2018.

η (%)	Institute/Company	Absorber	Year	Comment
22.9*	Solar Frontier (JAP)	CIGSSe	2017	1 cm <sup>2</sup> cell, with S, sequ. process
22.6*	ZSW (GER)	CIGSe	2016	
22.2 <sup>1</sup>	Solibro (SWE/GER)	CIGSe	2018	1 cm <sup>2</sup> cell
<b>22.0</b> <sup>2</sup>	AIST (JAP)	CIGSe	2018	after 25 h heat-light soaking
<b>20.9</b> <sup>3</sup>	ÅSC (SWE)	AgCIGSe	2017	1 cm <sup>2</sup> cell, CIGSe with Ag
20.8*	NREL (USA)	CIGSe	2014	
20.7	Toshiba (JAP)	CIGSe	2016	
20.4*	Empa (CH)	CIGSe	2013	on flexible polyimide foil
19.9	IEC Delaware (USA)	AgCIGSe	2015	CIGSe with Ag
19.7*	AGU (JAP)	CIGSSe	2013	with S, sequential process

\*) independently certified efficiency

1) O. Lundberg, presented at IW-CIGSTech 9 in Stuttgart, Germany (2018).

2) J. Nishinaga, T. Koida, S. Ishizuka, Y. Kamikawa, H. Shibata, and S. Niki, presented at E-MRS Spring Meeting 2018 in Strasbourg, France.

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Table 1. Power conversion efficiency η values of small-area Cu(In,Ga)Se<sub>2</sub> (CIGSe), (Ag,Cu)(In,Ga)Se<sub>2</sub> (AgCIGSe), and Cu(In,Ga)(S,Se)<sub>2</sub> (CIGSSe) chalcopyrite-type thin-film solar cells (see [1,2] and references therein; list is not complete).

#### **CIGSe** absorber modifications

Integral chemical composition and compositional gradings within the CIGSe absorber could have a significant influence on device performance. This relation is also relevant for different kinds of treatments of the absorber layer affecting bulk and surface properties of CIGSe prior to formation of the important CIGSe/buffer interface.

#### Improved Ga grading and increased Cu content

A method to increase  $J_{\rm sc'}$  mainly by increasing the absorption coefficient in the near-infrared (NIR) region, was described by Avancini et al. [3]. With the low-temperature CIGSe growth process established at Sharc25 partner Empa the [Ga]/([Ga]+[In]) (GGI) double gradient can be precisely controlled within the absorber, i.e. the increase of GGI to front and back side of the absorber. If the notch, the minimum of the GGI double grading, is widened and the Cu content is simultaneously increased, i.e. the [Cu]/([Ga]+[In]) ratio (CGI) raised to a maximum above a value of 0.90, a gain of approx. 1.1 mA/cm<sup>2</sup> could be achieved in  $J_{\rm sc}$  [3].

Numerical device simulations with the Sentaurus TCAD suite for different CGI and GGI profiles by the Sharc25 partner University of Parma support these experimental findings [4]. In addition, the increased light absorption in the NIR region by widening the notch could exactly be described by optical simulations developed at Empa calculating reflectance and external quantum efficiency (EQE) on the basis of experimental GGI depth profiles. The important dielectric functions of CIGSe and of the windows and back contact layers were determined





experimentally by a combination of ellipsometry, reflectance, and transmittance measurements [5].

### Post-deposition treatment (PDT) with alkali metals

Originally, the PDT of the CIGSe absorber with alkali metal salts was introduced by Rudmann et al. in 2004 [6]. NaF was thermally evaporated on the CIGSe front side after the CIGSe growth and afterwards annealed at 400°C to diffuse Na into the CIGSe absorber prior to CdS buffer growth by chemical bath deposition (CBD). This procedure is crucially required to provide the necessary Na content for high-efficiency solar cells if the CIGSe



Figure 2. Post deposition treatment (PDT) of CIGSe absorbers prior to buffer growth. a) Lowtemperature processed CIGSe on alkali metal-free substrates: First NaF- and in a second step KF-PDT is applied. b) High-temperature CIGSe growth on alkali-containing glass substrates: Only one PDT process is applied – KF-PDT (alternatively RbF- or CsF-PDT).



Figure 3. a) High-angle grain boundary (GB) at the atom probe tip prepared from a RbF-PDT treated high-temperature CIGSe absorber as revealed by transmission Kikuchi diffraction. b) Reconstruction of the same atom probe tip shows a Rb (K and Na) accumulation at the GB [14].

> absorber is deposited on a Na-free substrate – since Na usually inherently diffuses into the absorber during absorber formation at elevated temperatures when Na-containing substrates are used.

> A major step to push the CIGSe cell performance further was the discovery of the beneficial effect of potassium and the application to the front side of the CIGSe absorber in 2013 [7,8]. In this case the PDT was performed in-situ with KF (without breaking the vacuum) under a selenium atmosphere at temperatures typically around 350°C. This KF-PDT is applied after an initial NaF-PDT if an alkali-metal free substrate like polyimide, stainless steel foil or soda-lime glass with SiO<sub>x</sub> diffusion barrier is used as illustrated in Figure 2a. With this combined NaF-PDT + KF-PDT process, Empa was able to achieve a cell efficiency of 20.4% on polymer film using a lowtemperature CIGSe process [7].

> In the case of the high-temperature growth process, CIGSe is deposited onto Na- and K-containing glass substrate (soda-lime glass or alkali-aluminosilicate glass). These alkali metals diffuse from the glass substrates into the CIGSe absorber at elevated temperatures during the CIGSe process. In order to ensure the optimal amount of K in the CIGSe, a

KF-PDT is performed in a selenium atmosphere prior to CdS buffer growth by CBD (see Figure 2b).

The PDT method with alkali metals (or variations) was successfully adapted and applied by several research groups and companies worldwide leading to efficiency improvements on cell and module level. With the application of K on their chalcopyrite-based absorber layers, Solibro with a co-evaporation process and Solar Frontier with a sequential process, for example, have reported efficiencies of 21.0% [9] and 22.3% [10] (both with ARC) for chalcopyrite-based solar cells, respectively. Interestingly the K treatment works also for the penternary Cu(In,Ga)(S,Se)<sub>2</sub> absorbers with sulfur at the front side [11].

In 2016 a next step was taken by ZSW by introducing even heavier alkali metals like Rb and Cs in the PDT procedure. As a consequence, a new independently certified cell record efficiency of 22.6% with ARC was achieved. It should be noted that at ZSW CIGSe cell efficiencies above 20% with ARC could be achieved with all three PDT methods: KF-, RbF-, and CsF-PDT [12]. Many single cells were fabricated at ZSW during the Sharc25 project with high reproducibility and an efficiency level around 22% (with ARC). Also Empa improved their cell efficiencies for the lowtemperature process during the project duration approaching 21% efficiency using the double-PDT processes with RbF-PDT or KF-PDT after an initial NaF-PDT on both flexible polyimide foils and glass substrates with alkali diffusion barrier. Successful PDT treatment processes with higher alkali metal atoms like Cs was reported by Solar Frontier for sulfur-containing CIGSSe absorbers [13].

# Location of Rb in CIGSe, formation of defects, and alkali indium selenide phases

A statistical number of secondary ion mass spectroscopy depth profiles of CIGSe absorbers exposed to RbF- or CsF-PDT, showed that the intentionally introduced Rb (or Cs) was not only found at the CIGSe surface but was completely distributed inside the CIGSe absorber and was even observed at the Mo/CIGSe back side interface [12]. To investigate the location and distribution of Rb within the CIGSe absorber, highly spatially-resolved atom probe tomography (APT) measurements were performed by Sharc25 partner University of Rouen. Figure 3 shows an Rb accumulation on a nanometer scale at a high-angle grain boundary (GB) in a CIGSe absorber grown by the high-temperature process after RbF-PDT [14]. In addition, Na and K stemming from the alkali-containing glass are also enriched at the GB. The Rb concentration within the grain bulk is below the detection limit of the APT (10 ppm) whereas Na could be detected within the CIGSe grain bulk as well. The accumulation of Na at GBs in CIGSe was reported previously by Cadel et al. [15] and this property was suggested to passivate the GBs, which could be beneficial for the cell



performance. A comparison between an untreated sample without RbF-PDT and with RbF-PDT revealed that after Rb segregation at the GBs the Na concentration is reduced, suggesting that the lighter alkali element Na is replaced by the heavier element Rb at the GB [14].

The above described experimental finding (Rb at GBs in CIGSe measured by APT) is supported by the density functional theory (DFT) calculations performed by Sharc25 partner Aalto University. Based on a model for ternary CuInSe, with intrinsic bulk point defects and complexes [16] they theoretically investigated the insertion of alkali metal atoms from Li to Cs on different defect sites (interstitials, vacancies etc.) and the ensuing impurity migration in CuInSe, [17]. The formation energies and the vacancy mechanism migration barriers for alkali metal atoms on Cu sites are depicted in Figure 4. Li and Na in contrast to Rb and Cs have distinct effects on the structure of the CuInSe, absorber whereas K represents an interesting borderline case. Briefly, Li and Na could energetically be incorporated into CuInSe, grains, whereas Rb and Cs are more likely to accumulate at GBs and surfaces.

The stability of alkali metal phases was also studied, suggesting that metal phases Li<sub>x</sub>Cu<sub>i,x</sub>InSe<sub>2</sub> or Na<sub>x</sub>Cu<sub>i,x</sub>InSe<sub>2</sub>, i.e. Li and Na as impurities, might form at typical PDT substrate temperatures of 350°C and alkali metal concentrations <0.1 at% in CuInSe<sub>2</sub>. In contrast for heavier alkali metals like K, Rb, and Cs separated ordered AlkInSe<sub>2</sub> and CuInSe<sub>2</sub> phases are more probable to form [17]. Such a K-In-Se compound was experimentally evidenced using a combination of different surface sensitive techniques by Sharc25 partner Helmholtz-Zentrum Berlin for Materialien und Energie GmbH (HZB) for low-temperature processed CIGSe surfaces fabricated at Empa using various KF-PDT methods [18].

### Defects and barriers in high-efficiency CIGSe absorbers

In general, the CIGSe absorbers fabricated at Empa and ZSW exhibit no signatures of detrimental deep defects as measured with photoluminescence (PL) recorded at 10 K by Sharc25 partner University of Luxembourg [19], which were often reported by other groups. PL spectra of high-efficiency CIGSe absorbers often reveal interference fringes which negatively affect the detailed analysis of the PL spectra. These interferences can be overcome by angle-resolved PL measurements [20] and important parameters such as the quasi-Fermi level splitting could be extracted [21].

Admittance spectroscopy with an additional applied bias voltage by Empa revealed that at the interface-near CIGSe absorber region an additional capacitance step was observed which could be a result of the RbF-PDT procedure [22]. This new capacitance step at positive bias voltages is very likely caused by the PDT process (also with different types of alkali metals), which might form a transport barrier at the CIGSe front side for excessive alkali concentrations [23]. In contrast, the dominant capacitance step in conventional zero-bias admittance spectroscopy measured at University of Luxembourg appears to be independent of the alkali PDT and is best explained by an electron injection barrier located fully within the buffer/window stack [19].

Numerical simulations performed by Sharc25 partner University of Parma also explored the effect of several cell features and parameters on the capacitance-voltage characteristics and the apparent doping profiles extracted from them: these features and parameters include n-side doping densities, buffer thickness, and absorber/ buffer conduction band offset [24].

Figure 4. a) Bottom part of CuInSe, unit cell (Cu atoms in blue, In in purple, and Se in green) illustrating the mechanism of an alkali metal atom on a Cu site (Alk<sub>cu</sub> in red) exchanging to a Cu vacancy site (V<sub>cu</sub> in white). b) Formation energy of alkali metals Li to Cs on Cu site in CuInSe, (right vertical axis) and migration barrier for Alk<sub>cu</sub> - V<sub>cu</sub> and V<sub>cu</sub> - Alk<sub>cu</sub> mechanism (left vertical axis) after Malickaya et al. [17] (Adapted with permission from Ref. [17], ACS Publications. 2017).



Figure 5. Junction formation of CBD-CdS buffer layer on CIGSe subjected to RbF-PDT. a) Scheme of CBD-CdS buffer growth with different deposition times and estimated thicknesses on CIGSe after RbF-PDT. b) 2 keV hard X-ray photoelectron spectroscopy (HAXPES) measurement of CIGSe with RbF-PDT and solution-grown CdS on top. Different deposition times of CdS in CBD are indicated and the Rb-related HAXPES signals are highlighted by red arrows/dashed lines [25]. (Reproduced with permission from [25], ACS Publications, 2018).

### Influence of RbF-PDT on CIGSe surface and buffer growth by CBD

Na typically accumulates at the CIGSe surface after the CIGSe growth process and completely dissolves from the surface during the subsequent ammoniabased CBD of CdS. To investigate the situation in the case of RbF-PDT, CIGSe surfaces fabricated at high-temperature were analyzed at HZB after different deposition times of CdS (see Figure 5a) with synchrotron-based hard X-ray photoelectron spectroscopy (HAXPES) at

2 keV (corresponding to a maximal inelastic mean free path of approximately 4 nm). The HAXPES spectra depicted in Figure 5b clearly exhibit the Rb-related peaks which attenuate similar to the CIGSe-related Se 3s line with the growing buffer layer, i.e. the Rb is not dissolved by the CBD-CdS process [25]. The presence of a Rb-enriched surface was confirmed by lab-based X-ray photoelectron spectroscopy measurements performed at Empa on low-temperature CIGSe absorbers treated with RbF-PDT after different rinsing and etching procedures, similarly as observed for KF-PDT [26].

The RbF-PDT of CIGSe has also an influence on the growth of CdS buffer layer by CBD. Kelvin probe force microscopy (KPFM) measurements performed at Sharc25 partner INL revealed that the initial growth of CdS during the first few minutes leads to inhomogeneities in the surface and interface electronic properties. After about two to three minutes of CdS deposition a pn-junction forms, which was probed by a small surface photovoltage in some sample areas. After more than three minutes the surface photovoltage signal is large and homogeneous, indicating a complete junction formation [25].

## New approaches and concepts at CIGSe front side

The widely used CdS buffer layer in combination with the high resistive (HR) i-ZnO layer for CIGSe or CIGSSe solar cells and modules both absorb part of the short wavelength spectrum due to their bandgap energies of 2.4 and 3.2 eV, respectively, thus limiting  $J_{\rm sc}$ . In the last decades there were many efforts to substitute these layers with more transparent alternatives such as ZnS-based buffers or (Zn,Mg)O as HR layer. In addition, recombination losses at the CIGSe/buffer interface might occur, which can impair the device performance. Such losses could be mitigated by an optimization route borrowed from silicon solar cells. Employing a passivation layer in combination with point contacts at the front side of the CIGSe absorbers seems feasible and promising as discussed below.

#### Thinning of solution-grown CdS buffer layer

Within the Sharc25 project alternative buffer materials to the widely used CBD-CdS are under investigation, like solution-grown Zn(O,S) and mixed buffer layers like (Cd,Zn)S or ZnInS to increase transparency in the short-wavelength region. In addition, the consortium is working to thin the CBD-CdS down (i.e., decreasing its thickness) to a minimum to reduce detrimental absorption due to the CdS bandgap energy of 2.4 eV. This approach can be realized with different high resistive (HR) layers as alternatives to i-ZnO.

Figure 6 illustrates a CIGSe stacking sequence with the standard CBD CdS/i-ZnO buffer system on top of a CIGSe absorber without alkali metal PDT. The thickness of the CdS buffer layer is approximately 50 nm. Just reducing this thickness and still using i-ZnO would result in a decrease of the efficiency mainly due to reduced values of  $V_{\rm oc}$  and FF. If CIGSe with alkali PDT is used, the CBD-CdS buffer can be thinned down to 30 nm without any  $V_{\rm oc}$  losses. A further thickness reduction of CdS down to 20 nm is possible if sputtered (Zn,Mg)O is used as HR layer instead of i-ZnO. The CdS thickness can even be thinned down to 10 nm by using TiO, grown by atomic layer deposition (ALD) as alternative HR layer, resulting in an improved collection in the short wavelength region and thus increase in  $J_{sc}$  [27]. The standard TCO ZnO:Al was used for all stacking sequences described here. A further potential to increase efficiency is the combination with a TCO that offers higher mobility and thus enabling lower carrier concentrations and an improved IR transmittance. Such candidates could be hydrogendoped In<sub>2</sub>O<sub>2</sub> or InZnO which were successfully tested with different stacking sequences including CBD-Zn(O,S) buffers in combination with sputtered (Zn,Mg) O as HR layer [28].

## Passivation layers and point openings at CIGSe front side

The high cell efficiencies presented in Table 1 could be further improved by reducing recombination at the CIGSe/buffer interface. Typical approaches



Figure 6. Approach to thin the CBD-CdS buffer layer (from left to right) including the application of alkali metal PDT after CIGSe growth and the use of different HR layers like sputtered (Zn,Mg)O or ALD TiO, as an alternative for the commonly used sputtered i-ZnO.



Figure 7. a) Scheme of a CIGSe solar cell cross-section with an insulating passivation layer and a point contact opening between CIGSe absorber and CdS buffer layer. b) Calculated power conversion efficiencies  $\eta$  with 3D device simulation (Sentaurus TCAD) in dependence on point contact size (wpc) and pitch (d) [29]. c) SEM top view of point contact openings experimentally realized with hole-mask colloidal lithography on 14 nm thick ALD HfO<sub>2</sub> grown on polycrystalline CIGSe.

adopted from silicon solar cells are passivation layers applied to HIT (Heterojuntion with Intrinsic Thin layer) solar cells and structured point or line openings as they are implemented on the rear side of PERCs (Passivated Emitter and Rear Cell).

Figure 7a illustrates a CIGSe cross-section with an insulating passivation layer on top of CIGSe with point contact openings of a certain size and pitch, where CIGSe is in direct contact with the CdS buffer layer. Figure 7b shows the optimum size and pitch of point contacts on the CIGSe front side as calculated by 3D device simulations at the University Parma. To achieve a beneficial effect with a passivation layer and point contact openings at the CIGSe front side the size of the point contact should be in the range of several tens of nanometers, more or less independently of the pitch size [29].

Realizing such small openings on the relatively rough CIGSe surface (RMS roughness is typically around 100 nm) is a major challenge. Figure 7c shows a scanning electron microscope (SEM) top view image of polycrystalline CIGSe with typical grain sizes in the range of 1-2  $\mu$ m and a 14 nm thick ALD HfO<sub>x</sub> passivation layer on top. The openings were made with hole-mask colloidal lithography (HCL) by Sharc25 partner imec. The standard HCL method was specially optimized to CIGSe, but the opening sizes are in the range of 100- 200 nm, larger than the ideal simulated values of several tens of nanometers. Another challenge is the complete removal of the ALD passivation layer inside the holes to ensure optimum contact between the CIGSe absorber and buffer/TCO. Due to the large point contact openings applied to high-efficiency CIGSe cells, no increase in efficiency could be achieved with the passivation layers. Nevertheless, the successful application of the HCL method on the rough CIGSe front side could also be useful for other polycrystalline thin-film materials suffering from interface recombination losses, such as kesterite-type solar cells.

# Modifications at CIGSe back side and Mo/CIGSe interface

Similar to the CIGSe absorber front side, recombination losses can also occur at the Mo/ CIGSe back side interface. One solution could again be the application of a passivation layer in combination with point contacts, which were successfully employed for very thin CIGSe



Figure 8 a) Scheme of CIGSe absorber with GGI grading and standard Mo back contact. b) Scheme. of stacking sequence of CIGSe with Al back contact reflector with InZnO spacer and a very thin Mo or MoSe, layer on top. c) SEM cross-section image with InZnO spacer between Mo and CIGSe.



Figure 9: a) Atomic force microscopy (AFM) image of 10 nm thick ALD Al<sub>2</sub>O<sub>3</sub> with point contact openings with pitch size of around 2  $\mu$ m and point contact opening size of 200 nm realized by nanoimprint lithography on top of sputtered Mo back contact. b) AFM line profile extracted along red arrow in Figure 9a. layers [30]. With a non-graded CIGSe layer with thicknesses of 2-3 µm, these concepts can also increase efficiency. For point contact openings at the back side, calculations show that the size can be in the range of 200 nm and a typical pitch of around 2 µm would have a beneficial effect. Another approach to further enhance efficiency is to increase the optical path of photons within the CIGSe absorber layer with a back contact reflector or a mirror on top of the Mo contact. An advanced concept is a combination of such a reflector with a passivation layer and point contact openings.

#### **Back contact reflector**

An experimentally feasible approach to increase  $J_{\rm sc}$  in CIGSe solar cells by the use of a back contact reflector is illustrated in Figure 8. In this case a <100 nm thick

Al reflector is directly deposited on top of a standard substrate. To prevent diffusion of Al into CIGSe, which has a detrimental effect on cell performance, a 260 nm thick InZnO spacer is positioned between Al and CIGSe absorber as diffusion barrier. Even in the case of a 3 µm-thick CIGSe absorber grown by a low-temperature co-evaporation process at Empa there is an increase in NIR EQE and a pronounced subgap reflectance. The gain in  $J_{\rm sc}$  from reflection alone is around 0.3-0.7 mA/cm<sup>2</sup> and the best cell with reflector showed an efficiency of 19.9% compared to the best reference cell with 19.5% from the same CIGSe deposition run without reflector (both cells with MgF\_ARC) [31].

#### Back side passivation layer with point contacts

The atomic force microscopy (AFM) image in Figure 9a shows the realization of a 10 nm thick ALD Al<sub>2</sub>O<sub>2</sub> passivation layer with point contact openings on top of a sputtered Mo back contact. The passivation layer and point contact openings were fabricated by nano-imprint lithography (NIL) at imec and typical pitch between the openings is 2  $\mu m$  as shown in Figure 9b. So far, however, there has been no increase in efficiency through the use of the combination of a passivation layer and point contacts on the CIGSe back side for standard CIGSe thicknesses of 2-3 µm and standard double grading of GGI ratio. Nevertheless, the passivation effect could be proved by time-resolved PL measurements and the beneficial effect could be demonstrated for thin CIGSe films, similar to results reported by other groups.

#### Conclusion

The EU H2020 Sharc25 project has provided deep insights into highly efficient CIGSe thinfilm solar cells using advanced characterization methods, analytical tools, device simulation, and DFT modelling. This approach led to a continuous development and improvement of the CIGSe absorber, additional functional layers, and important interfaces. With optimized GGI gradient, CGI ratios, and alkali metal PDT with K, Rb, and Cs the efficiency of CIGSe thin-film solar cells prepared by the co-evaporation process (low- and high-temperature) could be significantly enhanced. The Rb intentionally inserted by PDT after the CIGSe growth accumulates at grain boundaries and is still present at the CIGSe front interface after the CBD process. It is very likely that AlkInSe, phases like RbInSe, form during the PDT process as calculated by ab-initio modelling. The  $J_{sc}$  of CIGSe cells could be increased by thinning down the CBD-CdS buffer layer in combination with a TiO, HR layer grown by ALD to reduce detrimental absorption in the UV. Another successful approach to enhance  $J_{sc}$ significantly was the implementation of an Al back reflector in combination with an InZnO spacer as diffusion barrier. Passivation layers grown by ALD on the front and back of the CIGSe absorber in combination with point contact openings were developed, but could not yet increase cell efficiency for CIGSe absorbers with standard thicknesses of 2-3 µm, as predicted by device simulations. Often an increase in one solar cell parameter is accompanied by a decrease in others. Nevertheless, during the Sharc25 project the efficiency values at Empa and ZSW were substantially increased and a value of 22.6% was achieved for a solar cell composed of a CIGSe absorber layer that underwent a RbF-PDT and featured a thinned CBD-CdS buffer layer, and a sputtered (Zn,Mg)O HR layer.

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